SUSTAINABLE PROCESS INTENSIFICATION USING BUILDING BLOCKS

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Abstract

In an immensely competitive market, many chemical plants need to increase their productivity for meeting the growing demands of chemical products, but at the same time, reduce the use of fresh raw materials and utilities to stay profitable. As the environmental regulations become stricter than before, they also need to reduce emissions and become environmentally sustainable. When resources are scarce, intensification and sustainability are the only choices for survival. However, there exists inherent trade-offs between the economics and the environmental sustainability of chemical process systems. In this work, we ask whether systematic process intensification can break through the current barriers posed by these trade-offs and lead to novel designs which are not only more economic, but at the same time, are also environmentally more sustainable compared to their non-intensified counterparts. Our recently proposed building block-based representation provides a systematic approach to analyze this, and we pose the sustainable intensification problem as a multi-objective MINLP optimization problem, which is solved using ε-constraint method. We demonstrate the methodology with a case study on ethylene glycol production and show that intensification could make chemical processes significantly more sustainable compared to the base case.

Keywords

Sustainable production, process intensification, process synthesis, multi-objective optimization.

Introduction

Most chemical plants are designed with concerns over techno-economic performance. Environmental regulations and sustainability metrics typically act as constraints on operational alternatives. However, as the demands for commodity products increase and the natural resources are depleted, sustainability considerations emerge as an important decision criteria in grass-root chemical plant design as well as for retrofitting.

A sustainable design is defined as an operation in which the consumption of resources "*do not exceed nature's capacity to provide the needed ecosystem goods and services*" (Bakshi, 2014). Besides the economics, the pursuit for a sustainable process design should also consider environmental and societal outcomes, which often times exhibit trade-offs between different design decisions. These trade-offs can be illustrated through optimal Pareto fronts, which describe the limits of our achievements for a given design task (Figure 1). This is illustrated through the Pareto curve I in Figure 1. While point A corresponds to optimal environmental footprint, it incurs a high cost in terms of process economics. Point B, on the other hand, corresponds to a design in which the cost is minimal, while it results in a high burden on process footprint. Traditional sustainable design methodologies strive for a '*middle ground*' between these extreme points in the presence of pre-determined

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alternatives (El-Halwagi, 2017; You et al., 2012; Carvalho et al., 2013).

However, a bottleneck in the search for more sustainable designs is often the lack of innovative flowsheets which can result in a shift towards a better Pareto front (e.g., Pareto Front II) with improvement in both the economic and sustainability metrics. Process intensification (PI) can be a potential vehicle towards such innovation through drastic improvements in process performance, e.g. cost, safety, volume and environmental footprint (Stankiewicz and Moulijn, 2000).

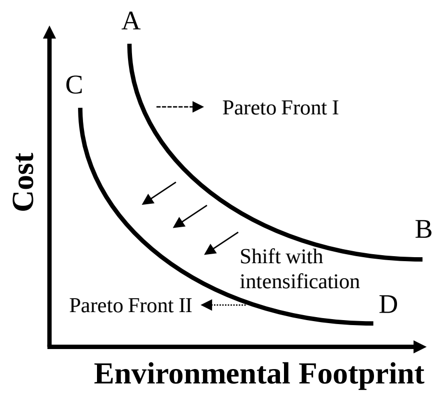

Figure 1. Example on the effect of process intensification on the Pareto front.

Identification of intensification alternatives and evaluating the effect of intensification requires systematic methodologies that can capture the synergy between different process phenomena, and, also analyze the trade-offs for the outcomes of intensification. Several systematic process intensification methods have been proposed in the past. These methods do not rely on unit operation-concept, rather employ more fundamental representations. Examples are generalized modular representation (Papalexandri and Pistikopoulos, 1996; Tian and Pistikopoulos, 2019), phenomena building blocks (PBB) (Lutze et al., 2013), elementary process functions (Freund and Sundmacher, 2008) and Infinite DimEnsionAl State-space (IDEAS) (Wilson and Manousiouthakis, 2000). A comprehensive review on these representations and methods can be found in Tian et. al. (2018) and Demirel et al. (2019). Phenomena building block-based approach developed by Gani and coworkers is also extended for sustainability considerations (Babi et al., 2015). An outstanding challenge is, however, to identify and analyze optimal intensification pathways for a given design problem while capturing the trade-offs between conflicting design decisions. To this end, our recently proposed building block-based approach (Demirel et al., 2017; Demirel et al., 2018; Li et al., 2018a, Li et al. 2018b, Li et al. 2018c; Li et al,, 2019) provides several unique advantages for process synthesis and intensification. Unlike traditional synthesis approaches that require superstructures with pre-postulated unit operation alternatives, our building block-based superstructure relies on physicochemical phenomena to automatically generate optimal intensified flowsheets. A mixed-integer nonlinear optimization (MINLP)-based model describes the superstructure. With this, novel intensified flowsheets can be generated through minimizing/maximizing several different objectives, e.g. economics, waste generation, utility consumption, etc. This results in a unique approach for process design and intensification in which the effects of intensification on the process economics as well as on the other sustainability metrics can be observed.

In this article, we demonstrate the benefits of this design approach in terms of sustainable process intensification. We first provide a brief overview on the building block-based representation. Then, we introduce the multi-objective model for building block-based design and demonstrate the benefits of this approach through an example on ethylene glycol production.

Building Block-based Process Intensification

Building block-based representation relies on an abstract building block definition which is used to represent many process phenomena, tasks, and equipment that can be used to come up with intensified/traditional process variants. These building blocks are characterized by their interior and surrounding four boundaries (borders). Mass and heat transfer associated with the material flows are facilitated through the streams flowing through these boundaries. Each building block has temperature, pressure and composition attributes and according to these attributes a phase is also assigned to the block. Temperature and composition of a block are determined through the material and energy balances around the block. A single or multiple building blocks can be used to represent many physicochemical phenomena (A detailed description can be found in Demirel et al. (2017)). To differentiate between different mass transfer operations, each boundary of a block is defined as either unrestricted, semi-restricted or completely restricted. If a boundary is unrestricted, then the flow though this boundary has the same composition with its source block. If it is semi-restricted, on the other hand, it indicates a mass transfer interception and its composition and flow rate is determined according to the nature of interaction between these two blocks. In vapor-liquid phase contact, for instance, phase equilibrium conditions dictate the value of these attributes. When these building blocks are combined in a two-dimensional grid, building block superstructure is obtained. Within this superstructure, different phenomena combinations yield many intensified/traditional equipment and flowsheet alternatives. An example on building block representation for an intensified reactive distillation process and its traditional reactor-separator-recycle counterpart are shown in Figure 2 along with the building block definitions used in representing these processes.

This grid-like superstructure representation also allows for a systematic mathematical formulation. The general optimization formulation for building block-based superstructure and how it can be utilized in a multi-objective optimization-based framework for sustainable process intensification is discussed in the following section.

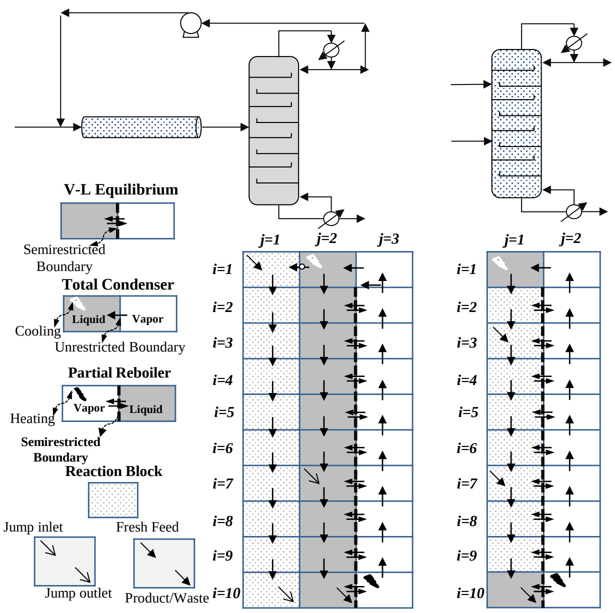

Figure 2. Building Block representations for standalone reactor-separator-recycle (left) and intensified reactive separation (right) system.

Mathematical Model for Sustainable Process Intensification

Each block within the building block superstructure can be identified with its position in the grid. Within a size of I × J grid, I representing the number of rows and J representing the number of columns, many different equipment and their interconnections can be represented within a single structure. To enable a systematic search for intensified flowsheet variants, a Mixed Integer Nonlinear Programming (MINLP) model is used where integer variables are used to determine the position of the physicochemical phenomena and enabling materials. The general single-objective MINLP describing building block superstructure is provided below (M1):

$$
\begin{aligned}
& \min \; f(x,y) && (1)\\
& s.t \quad g(x) = 0 && (2)\\
& \qquad q(y) \le 0 && (3)\\
& \qquad h(x,y) \le 0 && (4)\\
& \qquad\quad x^L \le x \le x^U \qquad x \in \mathbb{R}^N \;\; y \in \{0,1\}^M &&
\end{aligned}
$$

Here, x is a vector of continuous variables describing the material and energy flow associated with interblock streams, block temperature, pressure and composition, reaction rates, phase equilibrium constants, etc. y is a vector of binary variables that are used to determine the position of the active phenomena within the grid. Equation (1) is the objective function, Eq. (2) stands for the balance constraints which include material and energy balances. Equation (3) is for the assignment constraint which is a function of binary variables only and used for determining the position of the active material and phenomena assignments. Equation (4) stands for the logical constraints and they are used to relate continuous and binary variables. This MINLP model is used to generate flowsheet variants with binary variables indicating the position of active phenomena and material assignments within the grid. Furthermore, through fixing the position of several phenomena in a specific configuration, a fixed number of alternatives can be considered as is shown in Figure 2. This is also applicable for superstructure-based process synthesis with traditional/intensified units (Demirel et al., 2018).

The objective for the MINLP model can be related to process economics, environmental and safety criteria, and social benefits that can be accrued from the design. Each of these objectives may result in different optimal process configurations due to the trade-offs in between. To accommodate all these trade-offs, Eq. (1) needs to be updated as follows:

$$\min \; f_l(x,y) \qquad (5)$$

where $l \in \{1,\ldots,k\}$ represent different set of criteria that needs to be optimized. There are several methods available in addressing multi-objective optimization problems (Rangaiah, 2009). In this work, we will utilize ε-constraint method to determine the pareto optimal designs. In ε-constraint method, while one of the objective functions is kept, the others are converted to constraints. Hence, the multi-objective MINLP model describing sustainable process intensification through building blocks can be written as follows (M2):

$$
\begin{aligned}
& \min \; f_l(x,y) && (6)\\
& s.t \quad f_j(x,y) \le \epsilon_j \quad \forall\, j \in \{1,\ldots,k\} \setminus \{l\} && (7)\\
& \qquad Eqs. 3-4 &&\\
& \qquad\qquad x \in \mathbb{R}^N \;\; y \in \{0,1\}^M &&
\end{aligned}
$$

where ϵ_j represent the upper bound for the objective f_j, $j \neq l$. Solution of this model for a set of upper bounds yields the pareto front for the optimal block superstructure results. Next, we will demonstrate the use of this multi-objective optimization model for sustainable process intensification through building blocks-based approach with a case study on ethylene glycol production.

A Case Study on Ethylene Glycol Production

Ethylene glycol (EG) is used as an antifreeze in automobiles, desiccant for natural gas production, and a raw material for the production of polyester fibers and resins. Its industrial production is mainly based on the ethylene oxide (EO) hydrolysis:

Main: C_2H_4O (EO) + H_2O (W) → $C_2H_6O_2$ (EG)
Side: C_2H_4O (EO) + $C_2H_6O_2$ (EG) → $C_4H_{10}O_3$ (DEG)

Side reaction between EG and EO produces di-ethylene glycol (DEG). Heavier glycols, e.g. tri-ethylene glycol, are also possible via further side reactions which are neglected in this case study. The aim of the process is to produce 25 kmol/h EG with 95% purity. The same problem was addressed in Demirel et al. (2017) with building block superstructure for minimum utility consumption and a flowsheet suggesting the use of reactive and non-reactive V-L stages and two standalone reactors was obtained. This result suggest that reactive distillation can be a viable alternative for this process. Reactive distillation for EG production was also investigated by others with minimum utility cost and total capital cost objectives (Papalexandri and Pistikopoulos, 1996; Jackson and Grossmann, 2001). Here, we will perform a multi-objective analysis on two different design alternatives: (i) a non-intensified reactor-separator-recycle system which includes a plug-flow reactor (PFR) followed by a distillation column (DC), and (ii) an intensified reactive separation system which includes a single reactive distillation column (RD). The building block representations for these process alternatives are provided in Figure 2. In this particular case, we did not consider automatic flowsheet generation (but it is possible to do so). In representing plug flow reactor, 10 building blocks in series are used with the homogeneous reaction model provided by Altiokka and Karayalçin (2009). This corresponds to a 10-CSTRs-in-series model. For the separation columns, two building blocks, one in vapor and the other in liquid phase are used to represent an equilibrium stage. The number of building block pairs is a decision variable which stands for the optimal number of equilibrium stages. An upper bound of 50 is used for the number of building block pairs that can be used in separation including condenser and reboiler stages. The criteria for selection are the total annual cost (TAC) of the process (f_1) and CO_2-eq emissions from the operation (f_2) as a measure of the environmental impact of the design. While doing so, we also investigate the effect of intensification on the sustainability of the process.

Multi-objective Problem Formulations

The TAC of the process is evaluated based on the following objective function:

$$\begin{aligned} min\ f_l(x,y) = {} & \varphi\, Total\ Capital\ Investment \\ & + Utility\ Costs + Feed\ Costs \end{aligned} \tag{8}$$

while φ is the capital recovery factor (assumed as 0.33). Utility costs include hot utility, cold utility and electricity costs. Capital cost includes the PFR, column shell and tray, reboiler, condenser and recycle pump costs for reactor-separator-recycle system and reactive column shell and trays, reboiler and condenser costs for intensified system. Capital cost functions are obtained from Douglass (1988) and Jackson and Grossman (2001). Note that PFR cost is approximated as a multitubular heat exchanger with 1-in diameter tubes which facilitate fully developed turbulent flow regime through the reactor (Dye, 2001). Although there is no direct CO_2 emission from the process, three different sources of indirect emissions are identified and used in evaluating the environmental footprint of the process: CO_2-eq of the steam which is used as the hot utility, 0.0967 kg CO_2/MJ, CO_2-eq emissions related with the electricity production, 0.1541 kg CO_2-eq/MJ (Sheets and Shah, 2018), and CO_2-eq of the raw material use for EO, 163 kg CO_2-eq/kmol EO (Bergmann et al., 2007). CO_2-eq emissions from the production of EO includes emissions related to ethylene production. With these, the sustainability objective is formulated as follows:

$$min\ f_2(x,y) = E^{el} + E^{EO} + E^{steam} \tag{9}$$

Here, E^{el}, E^{EO} and E^{steam} are the CO_2-eq indirect emissions from the electricity, raw material EO and steam.

While using ε-constraint method, we choose TAC as the optimized objective and use sustainability objective as the constraint. First, optimal bounds on ε for the sustainability objective needs to be determined. The lower bound for ε is determined with Eq. (9) as the objective. This is performed for both flowsheet alternatives. Results from the solution of these problems correspond to the best designs in terms of sustainability. The upper bound for ε is determined with the following objective function:

$$min\ f_1(x,y) + \sigma f_2(x,y) \tag{10}$$

Here σ is a very small number (10^{-6}). Solution of the both flowsheets with Eq. (10) yields an upper bound on the ε parameter. We use 19 different ε values between the upper and lower bound and solve the resultant optimization problems which are in the form of model M2 with TAC as the objective. This procedure yields the pareto front for the two flowsheet alternatives. Because the number of discrete decisions are small, we can actually solve the MINLP problem for all combinations using separate NLPs. We used BARON (Tawarmalani and Sahinidis, 2005) to solve these NLPs and the optimality gap of the reported solutions are in the range of 20-23% for 12 hours of CPU time. The upper bound of the problem were often quickly found within few minutes while the improvements in the lower bound were rather slow.

Pareto Optimal Solutions

The pareto fronts for the two flowsheet alternatives are given in Figure 3. The x-axis shows the CO_2-eq emissions for per ton of EG product. The y-axis shows the TAC per ton of EG product. On this figure, while points A and B corresponds to the emission and cost optimal flowsheets for reactor-separator-recycle system, respectively, points C and D correspond to the emission and cost optimal flowsheets for the intensified flowsheet. These flowsheets are given in Figure 4 and a cost breakdown is provided in Table 1.

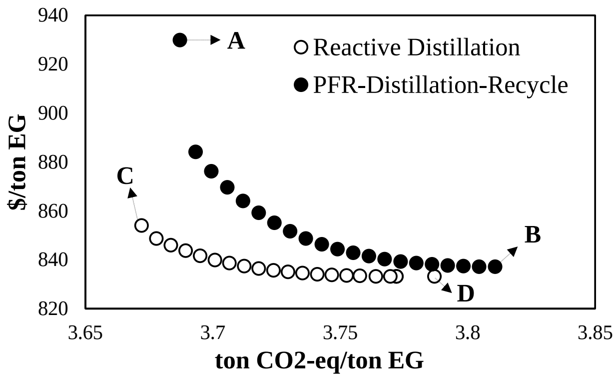

Figure 3. Pareto fronts for the reactive distillation and reactor-separator-recycle process.

Cost optimal intensified flowsheet (point D) features 19 active building blocks which correspond to a reactive column with 17 equilibrium trays, a partial reboiler and a total condenser. Column has a distributed feed structure and features a total reflux at the condenser. The cost optimal design for the reactor-separator-recycle system (point B) features a distillation column with 5 equilibrium trays, a partial reboiler and a total condenser. PFR operates adiabatically at high pressure. There is a large amount of recycle from the top of the column to the PFR and its water concentration is high. This helps into increasing the selectivity towards EG. There is nearly no reflux in the distillation column as the boiling points of EG and W differ significantly. When the cost optimal designs are compared, intensified design performs slightly better than the non-intensified flowsheet both in terms of cost (0.4%) and CO_2-eq emissions (0.6%).

Table 1. Cost of the optimal designs.

Pareto Point	A	B	C	D
Objective	Eq. 9	Eq. 10	Eq. 9	Eq. 10
Emissions (kton CO_2-eq/y)				
Total	54.63	56.46	54.41	56.10
From EO	39.45	39.41	39.45	39.37
From Steam	15.17	17.03	14.96	16.74
Cost ($MM/y)				
Capital	2.078	0.578	0.973	0.553
Hot utility	1.069	1.200	1.053	1.179
Cold Utility	0.053	0.058	0.052	0.057
Raw Material	10.578	10.566	10.578	10.557
TAC	13.778	12.405	12.656	12.346

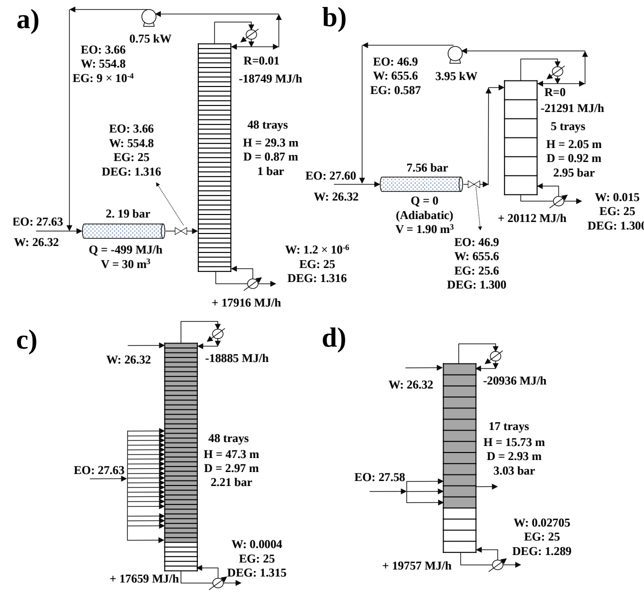

Figure 4. Flowsheets for the extreme pareto points.

When the objective is changed to Eq. (9), both flowsheets utilize all the allowed trays for the distillation columns (i.e. 48 trays). Reactive distillation process (point C) has again slightly lower emissions compared to non-intensified flowsheet (0.4%), yet it features much lower TAC (8.1%). In all four optimal results, the emission due to EO consumption has the highest share and it exhibits marginal changes among the optimal results. However, emissions from hot utility consumption differ significantly among the optimal results. (Note that emissions from electricity consumption and its effect on cost are negligibly small and not provided in Table 1).

Along the pareto fronts, changes are mainly driven by the increase in column height and PFR volume which result in reduced emissions from hot utility consumption at the expense of higher capital costs. The Pareto fronts show several interesting features. All the optimal points for the intensified flowsheet demonstrates less cost than the non-intensified counterpart for a given CO_2-eq emission value. This becomes more pronounced as we move towards the region with less emission. Hence, the intensified flowsheet not only has an improvement in the lowest emission possible from the process, but it also achieves this with significantly less cost.

Conclusions

In this work, we introduced sustainability considerations into the building block superstructure-based process intensification and synthesis methodology through multi-objective optimization. Here, we only demonstrated the applicability of the approach on two flowsheet alternatives for ethylene glycol problem. It has been observed that although intensification did not result in substantial decrease in the cost, it introduced significant benefits in terms of sustainability through a shift in pareto front. It also improved the lowest emission possible from the process with significantly reduced cost margin compared to its non-intensified counterpart. This highlights the importance of intensification in terms of sustainability and also multi-criteria evaluation techniques in the conceptual design of chemical processes. Note that this multi-objective formulation can be also utilized for automatic flowsheet generation and might bring further benefits in terms of systematic process intensification which will be demonstrated in a future contribution.

Acknowledgments

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